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Comment on "Rethinking first-principles electron transport theories with projection operators: The problems caused by partitioning the basis set" [J. Chem. Phys. 139, 114104 (2013)]

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I. INTRODUCTION

Reuter and Harrison (RH)[1] argue that a certain "decoupling condition" is required to be fulfilled in order to obtain correct results in transport calculations which employs an non-orthogonal basis (NOB) set, such as Ref. 2 and related in Ref. 1. The implicit assumption (decoupling condition) put forth by RH reads

$$\hat{N}_L \hat{H} \hat{N}_R = \hat{0}\,, \tag{1}$$

and violation is claimed to cause a "short circuit" problem. Here $\hat{N}_{L/R}$ are projection operators onto *L*/*R* regions defined in real-space, and $\hat{H}$ is the Hamiltonian operator describing the full system coupled via region *C* (see Fig. 1). RH state that their "short-circuit" problem resembles the ambiguity of assigning charge to atoms or regions in charge population analysis. Below we argue that this ambiguity problem does not carry over to calculations of charge flux.

We point out that Eq. (1) does not enter the NEGF derivations of current.[3] It is clear for infinitely separated *L* and *R* regions, we should be able to partition the system into scattering states originating from *L* and *R*, and thus Eq. (1) is, in principle, implicitly required to be asymptotically fulfilled. However, RH assign the *L* and *R* regions to the explicitly defined "physically motivated" regions in actual calculations. They attribute the "short circuit" to the fact that $H_{LR} = 0$ and $S_{LR} = 0$, for Hamiltonian (*H*) and overlap (*S*) matrices for a NOB set does not secure the operator equation above,

$$N_L S^{-1} H S^{-1} N_R = 0\,. \tag{2}$$

Several works address the NOB in transport calculations.[4–9] Here we provide simple arguments why equivalent OB and NOB sets yield the *same* transmission when the overlap is taken into account in the way discussed by Emberly and Kirczenow (EK).[4,10] We demonstrate by simple calculations how we get the exact results for transmission also when Eq. (2) is violated. Furthermore, we obtain the same transmission across an arbitrary dividing surface in real space as using a partitioning based on orbitals.

II. NONORTHOGONAL BASIS SET

We will argue that Eq. (2) can be violated while we obtain the exact transmission. First we consider a NOB chain-model with nearest neighbour hopping,[7] and overlap (matrix elements *t*, *s*). It can be shown that the range of the effective coupling, $|(S^{-1}HS^{-1})_{ij}| \propto ts^{|i-j|}$, is infinite, although rapidly decaying. This demonstrates how Eq. (2) is only possible as an asymptotic limit. On the other hand, the transmission can be calculated analytically *exact*[7] for this model by the EK method using a 1-site region for *C* thus explicitly violating Eq. (2).

Next, we consider an OB and an arbitrarily big central *C* region (Fig. 1). The transmission in terms of quantities defined inside *C*, reads[3,11]

$$T = \mathrm{Tr}[\bar{\Gamma}_L \bar{G} \bar{\Gamma}_R \bar{G}^\dagger]\,. \tag{3}$$

All matrices are given in the OB (denoted by a bar). Now we rotate the basis set to introduce an overlap matrix between the orbitals inside *C* using an "inverse" Löwdin transformation[12] from the OB to the NOB,

$$\begin{aligned} T &= \mathrm{Tr}[S^{\frac{1}{2}} \bar{\Gamma}_L S^{\frac{1}{2}} S^{-\frac{1}{2}} \bar{G} S^{-\frac{1}{2}} S^{\frac{1}{2}} \bar{\Gamma}_R S^{\frac{1}{2}} S^{-\frac{1}{2}} \bar{G}^\dagger S^{-\frac{1}{2}}] \\ &= \mathrm{Tr}[\Gamma_L G \Gamma_R G^\dagger]\,, \end{aligned} \tag{4}$$

where[10] $G \equiv S^{-1/2} \bar{G} S^{-1/2}$, $H \equiv S^{1/2} \bar{H} S^{1/2}$, and $\Sigma_{L/R} \equiv S^{1/2} \bar{\Sigma}_{L/R} S^{1/2}$.

We now split region *C* into C_1 and C_2. The range in the transport direction of the self-energies, Σ, in the NOB is that of *H* and *S*. For a big enough *C* and a NOB with finite range, we have zero matrix elements between *L*, 2 and 1, *R*. Thus Σ can be written as

$$\Sigma_L = \begin{pmatrix} (\Sigma_L)_{11} & 0 \\ 0 & 0 \end{pmatrix} \text{ and } \Sigma_R = \begin{pmatrix} 0 & 0 \\ 0 & (\Sigma_R)_{22} \end{pmatrix}, \tag{5}$$

which is the typical case, and we have

$$T = \mathrm{Tr}[(\Gamma_L)_{11} G_{12} (\Gamma_R)_{22} G_{21}^\dagger]\,. \tag{6}$$

We can write the GF in the NOB,

$$G^{-1} = \begin{pmatrix} g_{11}^{-1} & ES_{12} - H_{12} \\ ES_{21} - H_{21} & g_{22}^{-1} \end{pmatrix}, \tag{7}$$

where we introduce the inverse GF for region 1 without coupling to 2, $g_{11}^{-1} = ES_{11} - H_{11} - \Sigma_{11}$, and likewise g_{22}. It is

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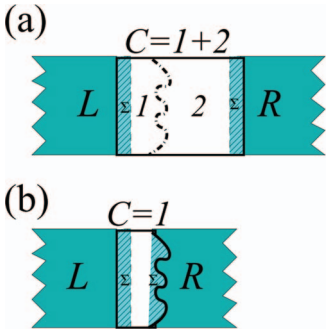

FIG. 1. The partition into left, central, and right regions used in the standard transport schemes. (a) Left and right self-energies are non-zero (in the nonorthogonal basis) in the dashed regions inside C. An overlap is included in region C. Region C is partitioned further into regions 1 and 2 to show how the transmission is unchanged when calculated using region 1 as a "new" central region (b), and the standard procedure for including overlap in transport calculations based on GFs.

straightforward from (7) to obtain

$$G_{11} = \left(g_{11}^{-1} - (ES_{12} - H_{12})g_{22}(ES_{21} - H_{21})\right)^{-1} \equiv \left(g_{11}^{-1} - (\widetilde{\Sigma}_R)_{11}\right)^{-1} \quad (8)$$

and

$$G_{12} = -G_{11}(ES_{12} - H_{12})g_{22}\,, \quad (9)$$

where we have introduced the right self-energy downfolded onto region 1,

$$(\widetilde{\Sigma}_R)_{11} = (ES_{12} - H_{12})g_{22}(ES_{21} - H_{21})\,. \quad (10)$$

Using $(\Gamma_R)_{22} = i[(g_{22}^{-1})^\dagger - (g_{22}^{-1})]$, we rewrite Eq. (6) and get a transmission formula for region $C = 1$ (Fig. 1(b)),

$$T = \mathrm{Tr}[(\Gamma_L)_{11}G_{11}(\widetilde{\Gamma}_R)_{11}G_{11}^\dagger]\,. \quad (11)$$

Thus we get *exactly* the same T for the NOB as for the original OB *if* we treat the overlap in the self-energy as Eq. (10), as is done in the "standard" EK approach.[4] We note that for the smaller region, $C = 1$, we may have high values of $|(S^{-1}HS^{-1})_{LR}|$ depending on S and H. But this is *not* relevant for the derivation.

In principle we can start by making a Löwdin transformation of the whole space. This leads to a long ranged H, but we may choose C big enough to ensure that $\bar{H}_{LR}$ is as small as we want. In actual calculations we make the C region small, as shown above, and need only to consider the range of H, S.

III. TRANSMISSION THROUGH A SURFACE DEFINED IN REAL-SPACE

The partition of space in terms of the orbitals for a given basis set yields the *same* result for T compared to a real-space division, when T is calculated with the same basis set. We sketch the proof and refer to Ref. 13 for details. The right flux normalized scattering states (label r) at energy, E, yield the current(T) through a surface, σ_{12}, located inside C,

$$T = \frac{\hbar}{m}\mathrm{Im}\sum_r \int_{\sigma_{12}} d\vec{\sigma}\cdot(\Psi_r^*(\vec{r})\vec{\nabla}\Psi_r(\vec{r})) \quad (12)$$

$$= \frac{\hbar}{m}\mathrm{Im}\sum_r \int_{V_1} d\vec{r}\;\Psi_r^*(\vec{r})\vec{\nabla}^2\Psi_r(\vec{r})\,. \quad (13)$$

We have rewritten the surface integral as an integral over region 1. In region 1 Ψ_r fulfils[11]

$$\left[-\frac{\hbar^2}{2m}\nabla^2 + V(\vec{r}) + \int d\vec{r'}\,\Sigma_L(\vec{r},\vec{r'})\right]\Psi_r(\vec{r'}) = E\Psi_r(\vec{r})\,, \quad (14)$$

where the integral is over the support of Σ_L. Note that σ_{12} is *arbitrary* as long as it is located within the freely chosen scattering region, and does not overlap with the support of Σ. Using Eq. (13) in Eq. (14) we can obtain

$$T = \frac{\hbar}{m}\mathrm{Im}\sum_r \int_{V_1} d\vec{r}\,(\Psi_r^*(\vec{r})\vec{\nabla}^2\Psi_r(\vec{r})) \quad (15)$$

$$= \frac{1}{\hbar}\sum_r \int d\vec{r}\int d\vec{r'}\;\Psi_r^*(\vec{r})\Gamma_L(\vec{r},\vec{r'})\Psi_r(\vec{r'})\,. \quad (16)$$

This is similar to the expression found by "embedding."[14] The main point here is the partitioning in terms of a basis set, $\{\phi_\alpha\}$, which overlap in regions of real space. We write

$$A_R(\vec{r},\vec{r'}) \equiv \frac{1}{\hbar}\sum_r \Psi_r^*(\vec{r})\Psi_r(\vec{r'}) = \sum_{r,\alpha\beta} c_{r\alpha}^* c_{r\beta}\,\phi_\alpha(\vec{r})\phi_\beta(\vec{r'}),$$
$$\Gamma_L(\vec{r},\vec{r'}) = \sum_{\alpha\beta}\phi_\alpha(\vec{r})(S^{-1}\Gamma_L S^{-1})_{\alpha\beta}\,\phi_\beta(\vec{r'})\,, \quad (17)$$

and using the definition of A_R we immediately obtain, $T = \mathrm{Tr}[\Gamma_L A_R] = \mathrm{Tr}[\Gamma_L G\Gamma_R G^\dagger]$, as we would when partitioning using the orbital basis set instead of σ_{12}.

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